Nuclear data requirements for the ADS conceptual design EFIT: Uncertainty and sensitivity study

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ARTICLE INFO

Keywords:
ADS
Cross-section uncertainties
Uncertainty propagation
Sensitivity technique
Monte Carlo simulation
Target accuracies

ABSTRACT

In this paper, we assess the impact of activation cross-section uncertainties on relevant fuel cycle parameters for a conceptual design of a modular European Facility for Industrial Transmutation (EFIT) with a "double strata" fuel cycle. Next, the nuclear data requirements are evaluated so that the parameters can meet the assigned design target accuracies. Different discharge burn-up levels are considered: a low burn-up, corresponding to the equilibrium cycle, and a high burn-up level, simulating the effects on the fuel of the multi-recycling scenario.

In order to perform this study, we propose a methodology in two steps. Firstly, we compute the uncertainties on the system parameters by using a Monte Carlo simulation, as it is considered the most reliable approach to address this problem. Secondly, the analysis of the results is performed by a sensitivity technique, in order to identify the relevant reaction channels and prioritize the data improvement needs. Cross-section uncertainties are taken from the EAF-2007/UN library since it includes data for all the actinides potentially present in the irradiated fuel.

Relevant uncertainties in some of the fuel cycle parameters have been obtained, and we conclude with recommendations for future nuclear data measurement programs, beyond the specific results obtained with the present nuclear data files and the limited available covariance information. A comparison with the uncertainty and accuracy analysis recently published by the WPEC-Subgroup26 of the OECD using BOLNA covariance matrices is performed. Despite the differences in the transmuter reactor used for the analysis, some conclusions obtained by Subgroup26 are qualitatively corroborated, and improvements for additional cross sections are suggested.

1. Introduction

One objective of the EU Integrated Project EUROTRANS (Eurotrans, 2005) was to accomplish a generic conceptual design of a modular European Facility for Industrial Transmutation (EFIT) (Artioli, 2006). This facility was aimed to demonstrate the feasibility and potential benefits of a dedicated Transuranium (TRU) burner. The potential benefits contributing to simplify the management of present and future radioactive waste include the large reduction of long term radioactivity, radiotoxicity and fissile materials inventories, as well as the minimization of short and medium term heat sources.

An adequate determination of such relevant parameters relies directly on the uncertainties of nuclear data. The subject of the impact of neutron cross-section uncertainties on the performance parameters of generic transmuter reactors, GEN-IV reactors, and their associated fuel cycles has been extensively studied in the last years, mainly by Aliberti et al. (2004) and Salvatores et al. (2008). Three main elements have to be defined to perform this kind of studies: (i) computational techniques enable to assess the impact of activation cross-section uncertainties on the prediction of relevant fuel cycle parameters, (ii) compilation of cross-section uncertainties and their correlation (variance–covariance matrices), and (iii) definition of the transmuter reactor characteristics and target accuracies.

Regarding the first point, the different uncertainty analyses performed are based on sensitivity theory (Aliberti et al., 2004, 2006). The most important limitations of this approach are: first, it is impractical to deal with the global effect of the complete set of

cross-section uncertainties; and second, the analysis based on a first order Taylor approximation does not allow accounting for non-linear effects and could fail when the nuclear data uncertainties are high.

Regarding the second point, most of performed uncertainty assessments have used a compilation of uncertainties known as ANL covariance data (Aliberti et al., 2004) (Palmiotti and Salvatores, 2005), and more recently, a new set of uncertainties known as BOLNA (Salvatores et al., 2008). ANL was a first compilation of "educated" guess of uncertainties, plausible but hypothetical; BOLNA covariance matrices are scientifically-based, but authors still recognize the preliminary nature of the data.

On the third point, the performed evaluations for ADS are based on a representative ADS-dedicated core with U-free, MA-dominated fuel on inert matrix, after one cycle irradiation (typically 1 year) (Salvatores et al., 2008).

It is recognized that new assessments are needed in the case of different ADS configurations, with different MA/Pu ratios and/or multiple recycle of the MA fuel. On the other hand, as more reliable uncertainty information becomes available, re-assessments of the impact on the integral parameters have also to be performed.

In the present study, an uncertainty and accuracy assessment is made for a "double strata" fuel cycle-based preliminary design of EFIT. The uncertainty data have been taken from the EAF-2007/UN data library (Forrest, 2007) since it includes data for all the actinides potentially present in the irradiated fuel (a lack of uncertainty information in the ANL/BOLNA can be found for nuclides beyond Cm^{247}). We use a Monte Carlo (MC) method to propagate the complete set of cross-section uncertainties to the isotopic inventory and related performance parameters. This method allows dealing with the overall/global effect of the complete set of uncertainties without any simplification. The applicability of this method for error propagation has been widely evaluated and applied by the authors in (Sanz et al., 2003) and more recently in (Sanz et al., 2006, 2007) within the frame of EUROTRANS-ADS applications. Monte Carlo methods are also being applied to propagate uncertainties of other kind of parameters, e.g. uncertainties in fundamental nuclear physics models (Koning and Rochman, 2008), showing the full potential of this *brute force* technique as computational power increases.

Notice that in this paper, we are concerned with the propagation of cross-section uncertainties on relevant fuel cycle parameters. In a future work, the impact of other sources of uncertainties, such as uncertainties in radioactive decay data and fission yield data, will be analyzed.

Section 2 is devoted to explain the main characteristics of the transmuter reactor and basic nuclear data used for calculations. In Section 3, the direct evaluation of uncertainties by using the MC technique is performed and the most contributing isotopes to each response function are shown. Once the uncertainties on the system parameters are evaluated, the next step is to analyze, by using a sensitivity methodology, the sensitivity profile of the parameters with respect to the cross sections, identifying the relevant reaction channels. This is done in Section 4. Then, after an optimization analysis, a table of cross section improvement requirements is given in Section 5, and a comparison with the accuracy requirements recently published by the WPEC-Subgroup26 of the OECD (Salvatores et al., 2008) using BOLNA covariance matrices is performed. Finally, conclusions include recommendations for future nuclear data measurement programs, beyond the specific results obtained with the present nuclear data files and the limited available covariance information.

2. Definition of the problem used in the analysis

The basic characteristics of the industrial-scale transmutation facility EFIT are: core cooled by pure lead, thermal power 400 MW, initial total mass of actinides 2.074 tonnes (21.7% MA, fuel initial composition shown in Table 1), and 150 GWd/tHM discharge burn-up corresponding to an equilibrium cycle (~778 irradiation days).

The problem of reference for us is to estimate uncertainties of the parameters of interest at the end of the irradiation period and along cooling time. In the calculations, a constant neutron environment representative of the equilibrium cycle is assumed for all the irradiation period (spectrum average energy $\langle E \rangle$ = 0.375 MeV and flux intensity 3.12×10^{15} n/cm^2 s). With this irradiation conditions, we reach a burn-up of 150 GWd/tHM at 778 days. The assumed neutron flux and spectrum have been taken from fully detailed 3D burn-up calculations performed with EVOLCODE2 code (Álvarez-Velarde et al., 2007), and correspond to a representative cell in the inner part of the core at mid-burnup.

In order to achieve the desired radiotoxicity reduction in a common double strata fuel cycle strategy (Von Lensa et al., 2008), multiple recycling is necessary, which means successive irradiation cycles inside the ADS. However, the uncertainty of the radionuclide content in the initial fuel within this scenario rises in each cycle since a high amount of the new fresh fuel is the result of previous irradiations with its corresponding uncertainty. Unfortunately, our method considers only the cross sections as uncertainty source but not the uncertainties existing in the fuel initial amount, as desired. We propose an alternative method to estimate as good as possible the final uncertainties in the isotopic content.

The ADS initial fuel in a cycle comes from: an 85% is the TRU reprocessed from the ADS irradiated fuel of the previous cycle; and a 15%, in our double strata strategy, comes from the first stratum (LWR) (we consider there is no uncertainty in the LWR isotopic content). Then, in a new ADS irradiation, although both fuel sources have been mixed in the fabrication process, the 85% of the fuel is propagating the uncertainties of the last cycle. After many cycles, the ADS fresh fuel would be propagating the uncertainties of previous cycles: a part of it is supposed to propagate uncertainties of only the previous cycle (150 GWd/tHM), other part of two cycles (300 GWd/tHM) and so on. On average, the ADS fresh fuel can be considered as a whole propagating the uncertainties coming from 3 to 4 previous cycles. Finally, we make the hypothesis that the uncertainties after the multiple irradiation period are very close to the uncertainties appearing in a single longer equivalent irradiation. For this reason, in our analysis, this effect is investigated considering single irradiation up to a discharge burn-up of 500 GWd/tHM (~3250 irradiation days). For these calculations, the neutron environment given above is assumed, since the fuel would be irradiated in the same reactor, characterised by a similar initial isotopic composition after having reached the equilibrium cycle.

The parameters to be evaluated are the ones selected as the most significant from the fuel cycle and the repository point of

Table 1
Initial actinide isotopic composition (#atoms). BOC indicates the beginning of cycle.

Isotope	BOC	Isotope	BOC
U-234	7.67E+25	Pu-244	1.55E+23
U-235	1.83E+25	Am-241	3.50E+26
U-236	2.54E+25	Am-242	3.81E+20
U-237	2.33E+18	Am-242m	2.96E+25
U-238	1.30E+23	Am-243	3.14E+26
Np-237	2.25E+26	Cm-242	3.17E+23
Np-238	6.07E+18	Cm-243	3.10E+24
Np-239	2.75E+20	Cm-244	2.67E+26
Pu-238	4.26E+26	Cm-245	7.82E+25
Pu-239	5.21E+26	Cm-246	5.20E+25
Pu-240	1.73E+27	Cm-247	1.12E+25
Pu-241	3.13E+26	Cm-248	8.33E+24
Pu-242	7.50E+26		

view in (Sanz et al., 2006). These are the actinide composition at the end of fuel cycle (to assess the transmutation potential), and related-response functions: decay heat, neutron emission and dose (radiotoxicity) by inhalation and ingestion at different cooling time (to assess the effect at the reprocessing, fuel fabrication and waste conditioning). The following years after discharge were selected as important for each response function: 0, 2, 50, 100, 200, 500, 1000, 10,000, 100,000 years after discharge for decay heat; 2, 3, 50 years after discharge for neutron emission; 0, 200, 500, 1000, 10,000, 100,000 years after storage for radiotoxicity.

Regarding target accuracies on the selected parameters, after discussion in different meetings in the frame of the IP-EUROTRANS project, it was finally decided that the values for the target accuracies are: 5% for the transmutation potential and 10% for all the other related parameters. These values are in agreement with those proposed in the recent bibliography (Aliberti et al., 2004, 2006).

2.1. Basic nuclear data used

The neutron cross-section uncertainty data have been taken from the EAF-2007/UN library, since it contains uncertainty information for all the actinides potentially present in the irradiated fuel. The EAF-2007/UN uncertainties seem to be conservative, though this library is based as much as possible of the experimental information. A thorough revision and extension of the uncertainty data for EAF-2010 has been performed (Kopecky, 2010), resulting in a new high-quality uncertainty file. Once this library becomes available, the EAF-2010 uncertainty data will be used for comparison and eventual update of the results obtained in this work.

EAF-2007/UN main characteristics are: (i) below 20 MeV, the energy spectrum is divided in three energy groups for nuclear reactions without threshold and in one-group for reactions with threshold; (ii) errors between all bands inside a certain energy group are 100% correlated, and errors between different energy groups are 0% correlated; and (iii) the uncertainty values stored in the library are $\Delta^2_{j,EAF}$ (j is the energy group), where $\Delta_{j,EAF}$ can be interpreted as the uncertainty (or relative error) in the standard or *best-estimate* cross section, stored in the corresponding standard activation library. For this analysis, the values of $\Delta_{j,EAF}$ are taken as three times the experimental uncertainty, that is, $\Delta_{j,EAF} = 3\Delta_{j,EXP}$.

To illustrate the uncertainties and large correlations implicit in this library, we show in Fig. 1 for the ^{241}Am(n,γ) reaction leading to ground state, the values stored in the library $\left(\Delta^2_{j,EAF}\right)$, the experimental uncertainty values taken for our calculations ($\Delta_{j,EAF}$) and the correlation matrix in the 15 multi-group energy structure used to produce both the ANL and BOLNA covariance matrices. It can be observed that uncertainties from EAF-2007/UN cause large positive correlations between the different energy bins included in a certain energy group.

To be consistent, since the objective of the present study is to provide a confident set of uncertainty estimates for EFIT, the associated best-estimated/nominal cross-section values have been taken from the EAF-2007 library. On the other hand, the required data to calculate the decay heat, radiotoxicity and neutron emission have been obtained processing different EAF-2007 basic libraries: decay data basic library, differential ranges for α-particles library, cross-section library for (α, n) reactions and commitment effective dose equivalent library.

3. Uncertainty evaluation by Monte Carlo approach

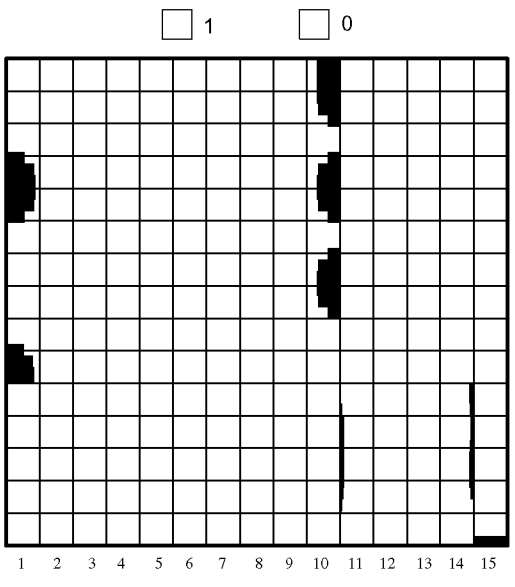
The inventory code ACAB (Sanz et al., 2008) is used to propagate the cross-section uncertainties on the relevant parameters by using a MC uncertainty technique. In this section, and for each selected parameter, we have proceeded as follows. First, the ACAB code has been used to calculate its nominal value (with no uncertainties) at the end of the two irradiation periods of interest (150 and 500 GWd/tHM). Then, ACAB has been run in the uncertainty mode to assess the impact of cross-section uncertainties, using a 1000 history-sampling. With the aim of identifying later the relevant reaction cross sections, the most contributing nuclides to the selected parameter are identified.

3.1. Inventory of actinides at the end of the irradiation periods

The nominal variation (no uncertainties considered) of the actinide concentrations at the end of the two irradiation periods is shown in Table 2 (columns 3 and 5 respectively). Uncertainty

Uncertainty values for Am^{241}(n,γ)

E_i (eV) - E_{i+1} (eV)	$\Delta^2_{j,EAF}$	$\Delta_{j,EXP}$ (%)
1.0E-05 - 1.0E-01	1.0000E-2	3.33
1.0E-01 - 1.5E+02	5.9487E-2	8.13
1.5E+02 - 2.0E+07	2.5000E-1	16.67



Gr	Energy
1	19.6 MeV
2	6.07 MeV
3	2.23 MeV
4	1.35 MeV
5	498 keV
6	183 keV
7	67.4 keV
8	24.8 keV
9	9.12 keV
10	2.03 keV
11	150 eV
12	22.6 eV
13	4.00 eV
14	0.54 eV
15	0.10 eV

Fig. 1. Uncertainty data found in the EAF-2007/UN for the ^{241}Am(n,γ) cross section leading to ground state and energy correlation structure implicit in the file.

Table 2
Initial actinide composition (N_i), nominal variation ($N_f - N_i$) and uncertainty (ratio between standard deviation and mean) in the final concentration (N_f) of the most relevant isotopes at different burn-up levels.

Nuclide	N_i (#atoms)	150 GWd/tHM		500 GWd/tHM	
		$N_f - N_i$ (#atoms)	Uncertainty (%)	$N_f - N_i$ (#atoms)	Uncertainty (%)
^{234}U	7.67E+25	−8.84E+24	4.64	−3.10E+25	16.05
^{235}U	1.83E+25	−8.50E+22	13.07	−4.25E+24	18.36
^{236}U	2.54E+25	−7.52E+23	1.75	−3.61E+24	7.61
^{237}U	2.33E+18	4.07E+22	7.91	3.59E+22	8.15
^{238}U	1.30E+23	−3.37E+21	1.29	−1.48E+22	4.98
^{237}Np	2.25E+26	−8.54E+25	6.27	−1.91E+26	23.68
^{238}Np	6.07E+18	2.40E+23	7.82	5.80E+22	9.98
^{239}Np	2.75E+20	2.92E+20	17.29	8.77E+18	12.43
^{238}Pu	4.26E+26	−2.69E+25	4.26	−2.05E+26	10.85
^{239}Pu	5.21E+26	−1.71E+26	4.57	−3.88E+26	12.94
^{240}Pu	1.73E+27	−2.84E+26	1.96	−9.23E+26	7.04
^{241}Pu	3.13E+26	−1.18E+25	8.17	−1.25E+26	14.66
^{242}Pu	7.50E+26	−7.31E+25	2.14	−2.83E+26	7.91
^{244}Pu	1.55E+23	2.80E+22	4.20	6.52E+22	9.78
^{241}Am	3.50E+26	−1.26E+26	7.15	−2.77E+26	20.75
^{242}Am	3.81E+20	1.31E+23	8.69	4.21E+22	8.35
^{242m}Am	2.96E+25	−1.15E+25	12.80	−2.48E+25	28.57
^{243}Am	3.14E+26	−3.60E+25	6.61	−1.24E+26	15.62
^{242}Cm	3.17E+23	2.61E+25	10.68	8.28E+24	7.70
^{243}Cm	3.10E+24	5.38E+23	23.25	−1.08E+24	32.61
^{244}Cm	2.67E+26	2.50E+25	6.02	5.20E+24	13.32
^{245}Cm	7.82E+25	−2.43E+24	13.34	−3.81E+24	18.76
^{246}Cm	5.20E+25	−8.67E+22	7.49	−3.05E+23	21.74
^{247}Cm	1.12E+25	−7.34E+22	15.38	−1.72E+23	27.18
^{248}Cm	8.33E+24	4.56E+23	6.40	1.49E+24	19.76
^{249}Bk	–	3.28E+23	19.19	4.19E+23	23.73
^{249}Cf	–	2.71E+23	19.59	8.08E+23	24.56
^{250}Cf	–	8.42E+22	31.90	4.56E+23	28.93
^{251}Cf	–	5.02E+21	42.57	8.84E+22	38.99
^{252}Cf	–	1.03E+20	52.36	5.53E+21	46.06

values (relative errors) of the final concentration computed by the MC approach are also given.

Results show the large uncertainties in the prediction of some actinides when using the EAF-2007/UN file, generally beyond design target accuracies. It is also found that the impact of the irradiation time on the uncertainties is very relevant. For the most important nuclides, uncertainties in concentrations increase almost linearly with the irradiation time. It is interesting to compare those values with the uncertainties computed by the WPEC-Subgroup26 of the OECD using BOLNA covariance matrices (hereafter called SG26) for ADMAB reactor, even if the image of the burner systems are different (Salvatores et al., 2008). For an equivalent burnup, uncertainties computed using EAF-2007/UN are of the same order of magnitude.

3.2. Decay heat along cooling time

The nominal values of the decay heat along cooling time, corresponding to both irradiation periods are in columns 2 and 4 of Table 3. The relative contribution of the most contributing actinides along cooling time is represented in Fig. 2; very similar percentages are found for the two irradiation periods. The major contributors before one year after discharge are the fission products; however, it can be seen that the presence of Cm increases the contribution of heavy isotopes already at short cooling times.

Results of the performed full uncertainty analysis of the decay heat using the MC technique are also provided in Table 3. Results show a relatively small impact of cross-section uncertainties on the decay heat, even as burn-up increases. That is, if a decay heat target accuracy of ±10% is assumed for future design studies, no substantial improvement of decay-heat-related cross sections would be needed. This conclusion corroborates results published by SG26 regarding to decay heat.

Table 3
Nominal values and uncertainties in the total decay heat along cooling time.

Time (years)	150 GWd/tHM		500 GWd/tHM	
	Nominal value (W)	Uncertainty (%)	Nominal value (W)	Uncertainty (%)
Shutdown	1.58E+07	3.63	9.27E+06	1.77
1	7.79E+05	4.68	5.42E+05	7.49
10	3.51E+05	4.02	2.93E+05	9.83
100	8.11E+04	2.37	4.73E+04	6.51
1000	1.10E+04	2.45	6.37E+03	5.76
10,000	2.62E+03	1.32	1.68E+03	4.38
100,000	2.55E+02	2.37	1.45E+02	6.24

3.3. Neutron emission along cooling time

An analysis of the neutron source has been performed, and main results are summarized in Table 4. In order to accurately compute the neutron emission due to (α, n) reactions, the methodology of the Los Alamos code SOURCES-4C (Wilson et al., 2002) was adopted and implemented in ACAB. Regarding to nominal values, a detailed analysis of the results (Álvarez-Velarde et al., 2009) has showed that neutron emission due to spontaneous fission (SF) is approximately one order of magnitude larger than due to (α, n) reactions. The major contributors to the SF neutron source are shown in Fig. 3. At low burn-up, the main contribution is due to Cm isotopes for all cooling times; however, as the burn-up increases, Cf isotopes become the major contributors (more than 75%) at short cooling times due to their larger amount. Regarding to (α, n) contribution, we have identified the Mg nuclides present in the fuel as the target materials responsible of the 99.9% of the (α, n) neutron production. If no Mg isotopes were present in the fuel, the (α, n) source would be dominated by reactions with C-13, O-17 and C-14, being Be-10 less important.

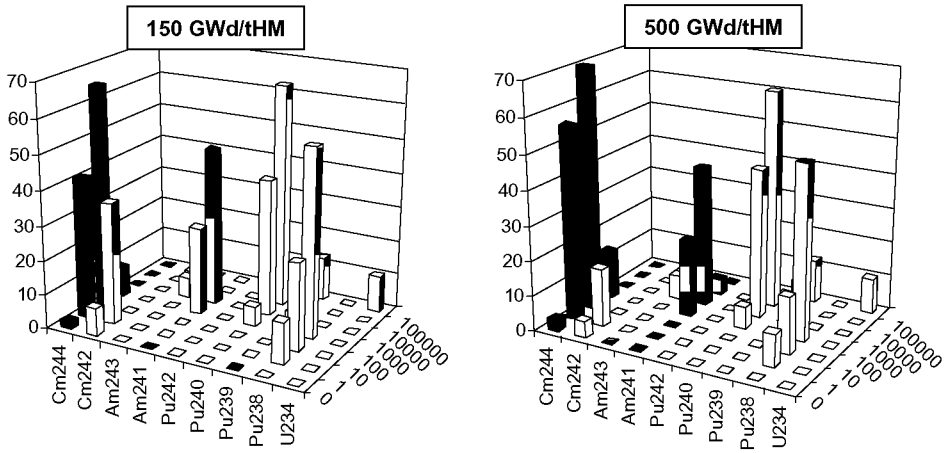


Fig. 2. Percentage contribution of the most contributing actinides to the total decay heat (at least 5% in one time step) along cooling time for different burn-up. Data table can be found in Álvarez-Velarde et al. (2009).

Table 4
Nominal values and uncertainties in total neutron emission along cooling time.

Time (years)	150 GWd/tHM		500 GWd tHM	
	Nominal value (n/s)	Uncertainty (%)	Nominal value (n/s)	Uncertainty (%)
Shutdown	3.63E+12	6.29	9.69E+12	29.22
1	2.57E+12	6.88	7.91E+12	28.14
2	2.27E+12	7.12	6.75E+12	26.41
3	2.15E+12	7.02	5.86E+12	24.59
50	6.32E+11	3.45	7.31E+11	9.81
100	4.03E+11	3.88	4.09E+11	11.88

With respect to the uncertainty analysis, a significant result is the strong impact of irradiation time on the uncertainty assessment: while uncertainties at 150 GWd/tHM are lower than the target accuracy of ±10%, they are not negligible as burn-up increases, due to a greater presence of heavier isotopes.

As far as neutron emission, results reported by SG26 do not consider contributions due to Cf isotopes, which can be not negligible even at low burnup, and might affect uncertainties in the neutron source.

3.4. Radiotoxicity along cooling time

To give an indication of the potential biological hazard of actinides, the committed effective dose equivalent (CEDE) by inhalation and ingestion has been computed and results are summarized in Table 5. Results show that the effective dose by inhalation is much larger than the effective dose by ingestion.

From Fig. 4, it can be seen that the main contributors to the dose by inhalation before one year of cooling time are ^{238}Pu, ^{242}Cm and ^{244}Cm for both irradiation periods. After one year, the contribution of isotopes such as ^{240}Pu and ^{241}Am increases. Very similar results can be found for CEDE by ingestion.

Regarding the uncertainty assessment, the present results confirm a relatively small impact of data uncertainties on the dose, even as burn-up increases. If a dose target accuracy of ±10% is required, no improvement of dose-related cross sections is needed.

3.5. List of relevant nuclides

In summary, it is found that generally the uncertainties on the nuclide densities at end of cycle increase with the irradiation period. When using the EAF-2007/UN data library, the computed uncertainties are significant and beyond design target accuracies for several isotopes even at low burnup. Results in response functions show a relatively small impact of data uncertainties on decay heat and radiotoxicity (uncertainties smaller than 10% at both irradiation periods). However, uncertainties in neutron emission can be significant as burn-up increases, exceeding the target value.

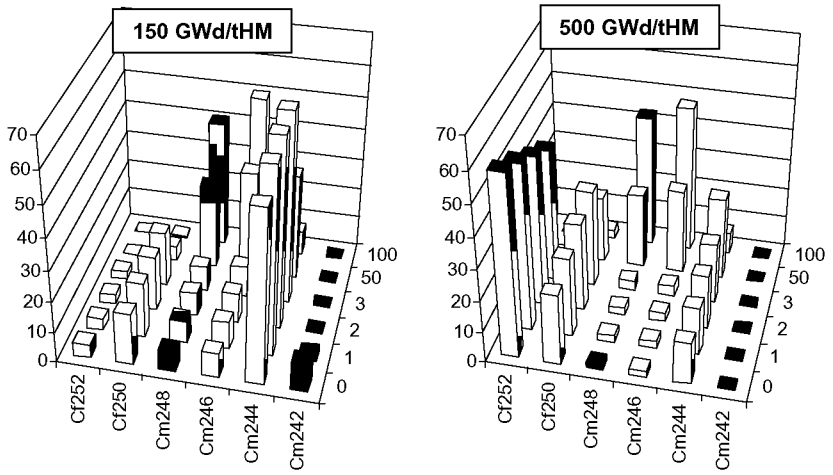


Fig. 3. Percentage contribution of the most contributing actinides to the spontaneous fission neutron source (at least 5% in one time step) along cooling time for different burn-up. Data table can be found in Álvarez-Velarde et al. (2009).

Table 5
Nominal values and uncertainties in effective doses by ingestion and inhalation along cooling time.

Time (years)	150 GWd/tHM				500 GWd/tHM			
	Nominal ING (Sv)	Unc. ING (%)	Nominal INH (Sv)	Unc. INH (%)	Nominal ING (Sv)	Unc. ING (%)	Nominal INH (Sv)	Unc. INH (%)
0	1.27E+12	3.87	4.37E+13	3.65	7.88E+11	1.96	3.01E+13	8.72
1	8.04E+10	3.34	3.61E+13	3.57	6.73E+10	7.76	2.68E+13	9.42
10	6.14E+10	3.17	2.82E+13	3.30	4.89E+s10	7.80	2.06E+13	8.94
100	1.92E+10	2.46	9.13E+12	2.50	1.09E+10	6.74	5.01E+12	7.09
1000	2.83E+09	2.09	1.34E+12	2.14	1.65E+09	5.25	7.79E+11	5.34
10,000	2.82E+09	2.80	3.63E+11	1.27	1.61E+09	6.67	2.29E+11	4.33
100,000	1.62E+10	3.72	1.39E+11	3.45	8.89E+09	9.36	7.70E+10	8.63

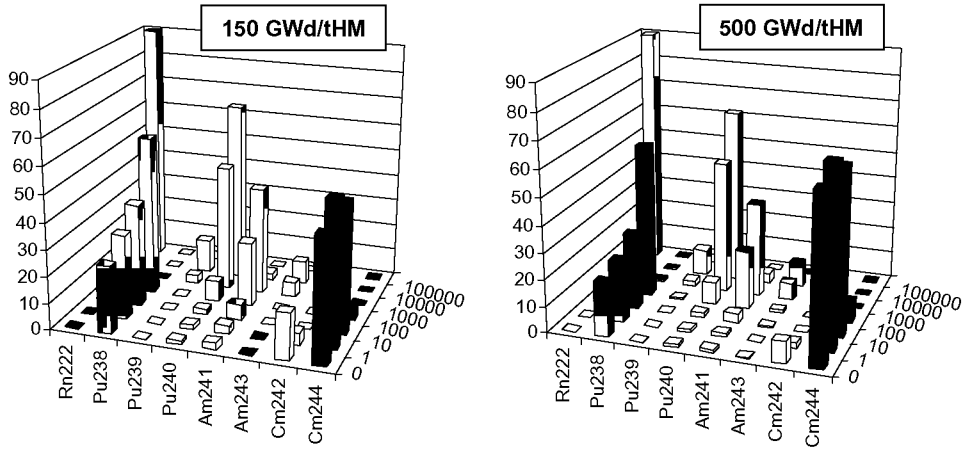


Fig. 4. Percentage contribution of the most contributing actinides to the dose by inhalation (at least 5% in one time step) along cooling time for different burn-up. Data table can be found in Álvarez-Velarde et al. (2009).

An inspection of Tables 2–5 and Figs. 2–4 leads to the list of the most relevant isotopes shown in Table 6, due either to their transmutation potential (T) or to their importance in the response functions (DH: decay heat, N: neutron emission, R: radiotoxicity).

4. Identification of critical cross sections

In order to identify the reactions whose cross-section uncertainties have a major impact on the concentration uncertainty of the actinides given in Table 6, sensitivity calculations have been performed.

Up to 31 cross sections have been identified as critical or potentially relevant, in the sense that their uncertainty causes a significant uncertainty in some of the isotopes of interest (uncertainty larger than the established target of 5%).

Table 7 shows the uncertainty in the concentration of each actinide-j due to the uncertainty in each potentially relevant cross section-i $\Delta N_j|_{\sigma_i}$ at the end of a 500 GWd/tHM burn-up cycle, as well as the full uncertainty (ΔN_j). Those uncertainty values refer to relative errors, which are computed as following:

$$\Delta N_j|_{\sigma_i} = \sqrt{(\rho_{ji}\Delta\sigma_i)^2} \quad \text{and} \quad \Delta N_j = \sqrt{\sum_{i=1}^{31}(\rho_{ji}\Delta\sigma_i)^2}$$

where ρ_{ij} is the sensitivity coefficient for the amount of nuclide-j due to the relative changes in cross section-i, and $\Delta\sigma_i$ is the relative error of the cross section-i.

Total uncertainties obtained with the sensitivity methodology at discharge can be compared with those obtained with the Monte Carlo methodology in Table 2. A very good agreement between both methodologies is found, demonstrating for this case the validity of the implicit linear approximation used in the sensitivity method.

Table 6
Most relevant nuclides and uncertainties in its concentration. (T: transmutation; DH: decay heat, N: neutron emission, R: radiotoxicity).

Nuclide	Uncertainty in concentration (%) Burn-up (GWd/tHM)		Relevant in			
	150	500				
U-234	4.6	16.1	T	DH		
U-235	13.1	18.4	T			
U-236	1.8	7.6	T			
Np-237	6.3	23.7	T			
Pu-238	4.3	10.8	T	DH		R
Pu-239	4.6	12.9	T	DH		R
Pu-240	2.0	7.0	T	DH		R
Pu-241	8.2	14.7	T			
Pu-242	2.1	7.9	T	DH		R
Am-241	7.2	20.7	T	DH		R
Am-242m	12.8	28.6	T			
Am-243	6.6	15.6	T	DH		R
Cm-242	10.7	7.7	T	DH		
Cm-243	23.3	32.6	T			
Cm-244	6.0	13.3	T	DH	N	R
Cm-245	13.3	18.8	T			
Cm-246	7.5	21.7	T		N	
Cm-247	15.4	27.2	T			
Cm-248	6.4	19.8			N	
Cf-250	31.9	28.9			N	
Cf-252	52.4	46.1			N	

The sensitivity coefficients of the response functions have also been determined from the sensitivity coefficients of the isotope concentrations. Then, the uncertainty (relative error) on the response function-F due to the uncertainty in each cross section-i will be given by $\Delta F|_{\sigma_i} = \sqrt{(\rho_{Fi}\Delta\sigma_i)^2}$, being the full uncertainty $\Delta F = \sqrt{\sum_{i=1}^{31}(\rho_{Fi}\Delta\sigma_i)^2}$, where ρ_{Fi} is the sensitivity coefficient for

Table 7
Uncertainties (in %) in the main actinides due to uncertainties in each cross section contributing with an uncertainty larger than 5% in at least one relevant nuclide. Sensitivity calculations correspond to the discharge, for a burn-up of 500 GWd/tHM.

		Isotope																			
		U^{234}	U^{235}	U^{236}	Np^{237}	Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{241}	Am^{242m}	Am^{243}	Cm^{242}	Cm^{243}	Cm^{244}	Cm^{245}	Cm^{246}	Cm^{247}	Cm^{248}	Cf^{250}	Cf^{252}
Uncertainty due to																					
U^{234}	(n,γ)	10.6	11.0	3.4	0.3																
	$(n,\gamma-M)$	10.5	10.9	3.4	0.3																
U^{235}	Fission		10.3	2.4	0.2																
Np^{237}	(n,γ)	0.4	0.3		23.4	0.6	0.8														
Pu^{238}	Fission	1.5	0.7			7.8	3.5	0.1													
	(n,γ)	1.3	0.7			7.1	6.3	0.5	0.3												
Pu^{239}	Fission						9.9	0.7	0.4		0.1										
Pu^{240}	(n,γ)			0.1		0.5	0.1	5.7	4.9	1.2	2.8	1.7	0.5	2.6	1.6	0.2					
Pu^{241}	Fission					0.9	0.2		13.0	2.2	5.7	3.1	0.9	5.1	2.8	0.3	0.1				
Pu^{242}	(n,γ)							0.2		6.5			6.5			4.1	2.9	0.8	0.4		
Am^{241}	(n,γ)	0.7	0.4		0.3	1.6	1.4			0.4	20.1	14.3	0.2	3.7	0.9	0.1	0.1				
	$(n,\gamma-M)$					0.4	0.1				1.8	13.0		1.6	1.1						
Am^{242m}	Fission					0.1						29.0	0.2	0.4	0.3	0.1					
	(n,γ)											6.0	0.3			0.2	0.2				
Am^{243}	$(n,\gamma-M)$							0.3	0.2				13.5			3.8	4.2	1.8	1.1	0.2	
Cm^{242}	(n,γ)	0.1				0.6	0.2							1.4	27.5						
Cm^{243}	Fission														16.3						
	(n,γ)														6.2						
Cm^{244}	(n,γ)							0.6	0.3						0.1	11.2	14.9	8.0	5.4	1.2	0.3	–
Cm^{245}	Fission																7.9	3.5	2.2	0.5	0.1	–
	(n,γ)																5.7	16.2	12.1	3.3	1.1	0,4
Cm^{246}	(n,γ)																	11.6	18.2	8.7	4.3	2,3
Cm^{247}	Fission																	–	12.1	5.0	2.3	1,2
	(n,γ)																		6.9	14.7	9.3	6,3
Cm^{248}	(n,γ)																			5.1	16.2	17,3
Bk^{249}	(n,γ)																				14.4	17,0
Cf^{249}	(n,γ)																				10.9	9,4
Cf^{250}	Fission																				5.4	3,8
	(n,γ)																				6.8	24,6
Cf^{251}	Fission																					13,0
																						27,9
Total		15.5	19.0	7.5	23.7	10.8	13.2	7.4	14.6	8.1	21.3	35.8	15.3	7.6	32.8	13.0	18.7	22.0	26.6	19.0	27.9	48.0

the response function-F due to the relative changes in cross section-i.

In Table 8, the uncertainties in the response functions for a burn-up of 500 GWd/tHM are given. The cooling times represented for each response function are: 100 years for decay heat (DH), 2 years for the neutron emission (N), and 10,000 years for the radiotoxicity by inhalation (INH).

Concerning the critical cross sections, the following issues are to be remarked:

- There is no $(n, 2n)$ cross sections with significant effects.
- (n, γ) Cross sections of ^{242,244,245,246,247}Cm, ^{237}Np, ^{241}Am cause very significant effects, leading to an uncertainty in the concentration of some relevant actinide larger than 14%. Additionally, captures of ^{248}Cm, ^{249}Bk, ^{250,251}Cf have also very significant effects, in this case, for the neutron source prediction. Smaller impact has capture of ^{234}U, ^{238,240,242}Pu, ^{242m}Am, ^{243}Cm.
- Capture to the metastable state cross sections of ^{234}U and ^{241,243}Am are relevant for the prediction of ^{234}U, ^{235}U, ^{242m}Am, ^{243}Am.

Table 8
Uncertainties (in%) in response functions due to uncertainties in each cross section contributing with an uncertainty larger than 0.5%. Sensitivity calculations correspond to the discharge, for a burn-up of 500 GWd/tHM. (DH-100y: decay heat after 100 years cooling time, N-2y: neutron emission after 2 years cooling time, INH-10,000y: CEDE by inhalation after 10,000 years cooling time).

		Response function		
		DH–100y	N-2y	INH-10,000y
Uncertainty due to				
Np^{237}	Fission	0.53		
	(n, γ)	0.55		0.28
Pu^{238}	Fission	1.98		0.51
	(n, γ)	1.90		0.75
Pu^{239}	Fission	0.39		0.93
	(n, γ)	0.53		0.94
Pu^{240}	Fission	0.93		1.50
	(n, γ)	1.28		1.72
Pu^{241}	Fission	1.78	0.28	0.50
	(n, γ)	0.90	0.34	0.62
Pu^{242}	Fission	0.34	0.33	0.62
	(n, γ)	0.89	0.91	1.44
Am^{241}	Fission	0.71		
	(n, γ)	1.43		0.44
	$(n, \gamma - M)$	0.63		
Am^{242m}	Fission	0.70		0.27
	(n, γ)	0.37		0.32
Am^{243}	Fission	0.40	0.40	0.64
	(n, γ)			0.47
	$(n, \gamma - M)$	0.92	0.90	1.78
Cm^{242}	Fission	0.41		
	(n, γ)	0.64		
Cm^{243}	Fission			
	(n, γ)			
Cm^{244}	Fission	0.74	0.80	0.91
	(n, γ)	1.42	1.61	1.90
Cm^{245}	Fission		0.40	0.84
	(n, γ)	0.32	1.00	0.93
Cm^{246}	Fission		0.41	0.27
	(n, γ)		1.68	0.57
Cm^{247}	Fission		1.14	
	(n, γ)		2.43	
Cm^{248}	Fission		1.00	
	(n, γ)		3.56	
Bk^{249}	(n, γ)		3.46	
Cf^{249}	(n, γ)		2.73	
Cf^{250}	Fission		1.81	
	(n, γ)		3.66	
Cf^{251}	Fission		2.46	
	(n, γ)		3.61	
Cf^{252}	Fission		1.15	
	(n, γ)		0.91	
Total		7.3	28.5	6.8

- Fission cross sections of ^{242m}Am and ^{243}Cm have very significant effects for the isotope prediction of ^{242m}Am and ^{243}Cm respectively. Fission of ^{235}U, ^{238,239,241}Pu, ^{245,247}Cm as well as fission of ^{250,251}Cf (important for the neutron emission) are less relevant.

Let us compare Table 7 with the uncertainty breakdown on the nuclide densities computed by SG26 (from Np^{237} to Cm^{246}) for ADMAB reactor. Values of the sensitivity coefficients are logically very different, since the initial actinide composition, spectrum and burn-up differ from our problem. However, some interesting conclusions can be drawn when comparing the cross sections responsible of the overall uncertainty of each nuclide.

- We identify the same capture and fission cross sections as responsible of the Np^{237} uncertainty.
- The major contributions to the uncertainties in Pu isotopes are due to the same capture and fission cross sections. We identify besides small contribution of other reactions, but causing an uncertainty lower than 1.5% in any Pu isotope.
- In the case of Am isotopes, we have found that apart from the capture and fission cross-sections identified by SG26, there are other relevant data sources: ^{241}Am$(n, \gamma - \mathrm{M})$ for the overall uncertainty of ^{242m}Am, and ^{243}Am$(n, \gamma - \mathrm{M})$ for the overall uncertainty of ^{243}Am.
- In the case of Cm isotopes, in addition to the reactions identified by SG26 as the major data sources, we have found significant the impact of other cross sections: for the overall uncertainty of ^{242,243}Cm, fission of ^{241}Pu and capture of ^{240}Pu; for the overall uncertainty of ^{244,245}Cm, capture of ^{242}Pu and $(n, \gamma - \mathrm{M})$ of ^{243}Am.

We have also compared Table 8 and the uncertainty breakdown on the decay heat at 100 years after discharge and neutron source at 2 years after discharge computed by SG26 for ADMAB reactor. The most important difference is the contribution of cross sections of isotopes beyond Cm^{245} to the neutron source. Whereas such cross sections are not present in SG26 results, our analysis indicates a strong impact of capture cross sections of $^{246\text{-}247\text{-}248}$Cm, ^{249}Bk as well as capture and fission reactions of some Cf isotopes.

5. Table of required accuracies

Cross-section uncertainties can be reduced so that the selected integral parameters fulfill the design target accuracies. With the aim of evaluating the reduction level and establishing priorities when using the EAF-2007/UN uncertainty data library, an optimization problem is solved. Results of this optimization have to be understood in the frame of the used uncertainty library, the taken ADS fuel composition and the selected target accuracies.

The following objective function is minimized, using the solver DONLP2 (Spellucci, 1998), based on a SQP method:

$$\sum_{i=1}^{N} \lambda_i / x_i^2, \quad i = 1, \ldots, N$$

where $x_i = \Delta_{1Gi}$ (uncertainty in one-group of the i reaction), N is the total number of reactions whose cross-section uncertainties are to be determined and λ_i represents a cost parameter related with each cross section (in our case, we take a constant value $\lambda_i = 1$). The objective function is constrained to the following boundary conditions: (i) $x_i \geqslant 0$; (ii) the maximum uncertainty in the concentration of all the nuclides of interest must be lower than the target uncertainty of 5%; and (iii) the maximum uncertainty in the response functions in the cooling times in which each response function is relevant must be lower than the target value of 10%.

Table 9
One-group uncertainties in the critical cross sections processed from EAF-2007/UN (Δ_{EAF}), uncertainties required for satisfying the target accuracies (Δ_{target}) and required uncertainty reduction ($\Delta_{EAF}/\Delta_{target}$). Calculations correspond to a burn-up of 500 GWd/tHM.

Reaction		Δ_{EAF}	Δ_{TARGET}	$\Delta_{EAF/DTARGET}$
U^{234}	(n,γ)	38.9	7.1	5
	$(n,\gamma-M)$	38.9	7.1	5
U^{235}	Fission	12.9	4.2	3
Np^{237}	(n,γ)	14.3	2.8	5
Pu^{238}	Fission	12.3	6.4	2
	(n,γ)	14.5	5.2	3
Pu^{239}	Fission	9.6	3.4	3
Pu^{240}	(n,γ)	9.3	4.8	2
Pu^{241}	Fission	15.6	4.2	4
Pu^{242}	(n,γ)	12.6	5.3	2
Am^{241}	(n,γ)	15.8	2.8	6
	$(n,\gamma-M)$	15.9	2.9	5
Am^{242m}	Fission	24.0	2.4	10
	(n,γ)	32.8	6.2	5
Am^{243}	$(n,\gamma-M)$	15.3	4.1	4
Cm^{242}	(n,γ)	30.0	3.4	9
Cm^{243}	Fission	16.0	3.2	5
	(n,γ)	32.0	7.4	4
Cm^{244}	(n,γ)	24.6	4.6	5
Cm^{245}	Fission	9.7	4.1	2
	(n,γ)	32.7	5.5	6
Cm^{246}	(n,γ)	28.2	4.3	7
Cm^{247}	Fission	16.5	4.0	4
	(n,γ)	32.1	5.0	6
Cm^{248}	(n,γ)	19.2	2.5	8
Bk^{249}	(n,γ)	31.7	3.2	10
Cf^{249}	(n,γ)	32.4	4.3	7
Cf^{250}	Fission	33.0	6.9	5
	(n,γ)	29.3	2.6	11
Cf^{251}	Fission	31.6	3.7	9
	(n,γ)	29.9	2.4	12

From results in Table 9, the following list of priorities can be deduced:

- Fission cross sections of ^{242m}Am, ^{243}Cm, ^{250,251}Cf.
- (n,γ) of ^{234}U, ^{237}Np, ^{241,242m}Am, ^{242,244,245,246,247,248}Cm, ^{249}Bk, ^{249,250,251}Cf.
- $(n,\gamma-M)$ of ^{234}U, ^{241}Am.

Notice that this table of target accuracies is the required uncertainty reduction for the diagonal + correlation current uncertainties implicit in the EAF-2007/UN library. That is, the table is only as valid as the EAF-2007 data library. The use of different uncertainty libraries, with different relative errors, energy correlation structures or with cross-correlations among reactions, would lead to considerably different predictions of uncertainties in the isotopic concentration and associated functions; then, the list of priorities could be significantly different.

6. Conclusions

Given a cross-section uncertainty data library, we are able to determine how uncertainties might affect the fuel cycle parameters for a given system. And we could find the relevant reaction channels as well as prioritize the data improvement needs.

An uncertainty analysis of the impact of activation cross-section uncertainties on the main fuel cycle related parameters has been performed for a preliminary design of EFIT, using the EAF-2007/UN uncertainty data library. We recommend the Monte Carlo technique as the suitable tool to compute the effect of the complete set of uncertainties on the parameters, and the sensitivity technique as the suitable tool for the analysis phase.

Results show that generally, uncertainties increase very significantly with the irradiation period. Uncertainties on the nuclide densities at end of cycle are significant (larger than the considered target value of 5%) for several actinides and would not be acceptable in a final EFIT design. A relatively small impact of cross-section uncertainties on the decay heat, neutron emission and radiotoxicity is seen at short burn-up (lower than 10% along cooling time); at higher burn-up, the neutron emission could not meet the target accuracy requirements.

Consequently, it is necessary to improve the quality of some nuclear data. By means of a sensitivity technique, a set of 31 reaction channels have been identified as critical for the actinide isotopic prediction and related-response functions. A target accuracy assessment has been performed, leading to a priority list for new evaluations. However, as this specific list could change significantly depending on the selected target accuracies and the used uncertainty data, the focus must be on the sensitivity results. They lead to conclusions nearly independent on the basic library used and, consequently, they are of a major general interest. For practical programming of new nuclear data activities, it is important to address the isotopes and cross sections with significant sensitive coefficients and uncertainties.

In this sense, the present study allows recommending for future nuclear data measurement programs, improvements for the following cross sections:

- Concerning minor actinides, significant improvements are required for (n,γ) cross sections of nearly all the isotopes of Cm, ^{237}Np, ^{241}Am, ^{249}Bk, and ^{250,251}Cf, as well as for $(n,\gamma-M)$ cross sections of ^{241}Am, ^{243}Am. Strict requirements are also found for fission of ^{242m}Am and ^{243}Cm. Smaller uncertainty reductions would be required for capture of ^{242m}Am, ^{249}Cf, and fission of ^{245,247}Cm, ^{250,251}Cf.
- Concerning the major actinides, uncertainty reduction is needed for fission of ^{235}U, ^{238,239,241}Pu, for (n,γ) of ^{234}U, ^{238,240,242}Pu and $(n,\gamma-M)$ of ^{234}U.

When comparing our results with evaluations performed by WPEC-Subgroup26 of the OECD using BOLNA covariance matrices, several conclusions can be drawn, despite the differences in the transmuter reactor characteristics used for the analysis. A first relevant conclusion is the impact of the uncertainty data on the uncertainty assessment. The different uncertainty prediction, the different selected target accuracies, as well as the study of other integral parameters (criticality, power peak value, ...) leads to different list of nuclear data priorities. A second important issue is that the cross sections found to be relevant for actinide density, decay heat and neutron source predictions in ADS designs are basically the same, although our analysis suggests the need of improvement of additional cross sections (capture and fission of some isotopes beyond Cm^{247}, important for the neutron source prediction at fuel fabrication).

Acknowledgements

This work is partially supported by the CIEMAT-ENRESA collaboration and by the European Union IP-EUROTRANS-NUDATRA Domain.

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